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Surface studies of liquid Li and Sn-Li

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outline

- Background
- The experiment
- Data for liquid Li
- Data for liquid Sn-Li alloy
- Summary and plans



Surface studies for plasma-facing liquids

Issue: What comprises the surface seen by the plasma?

Need: Fundamental data on liquid surfaces needed to properly model conditions that will exist in a fusion reactor.

Task: Examine surface composition of candidate liquids:

- what is impurity content?
- how does composition change with temperature?
- do segregation effects occur?

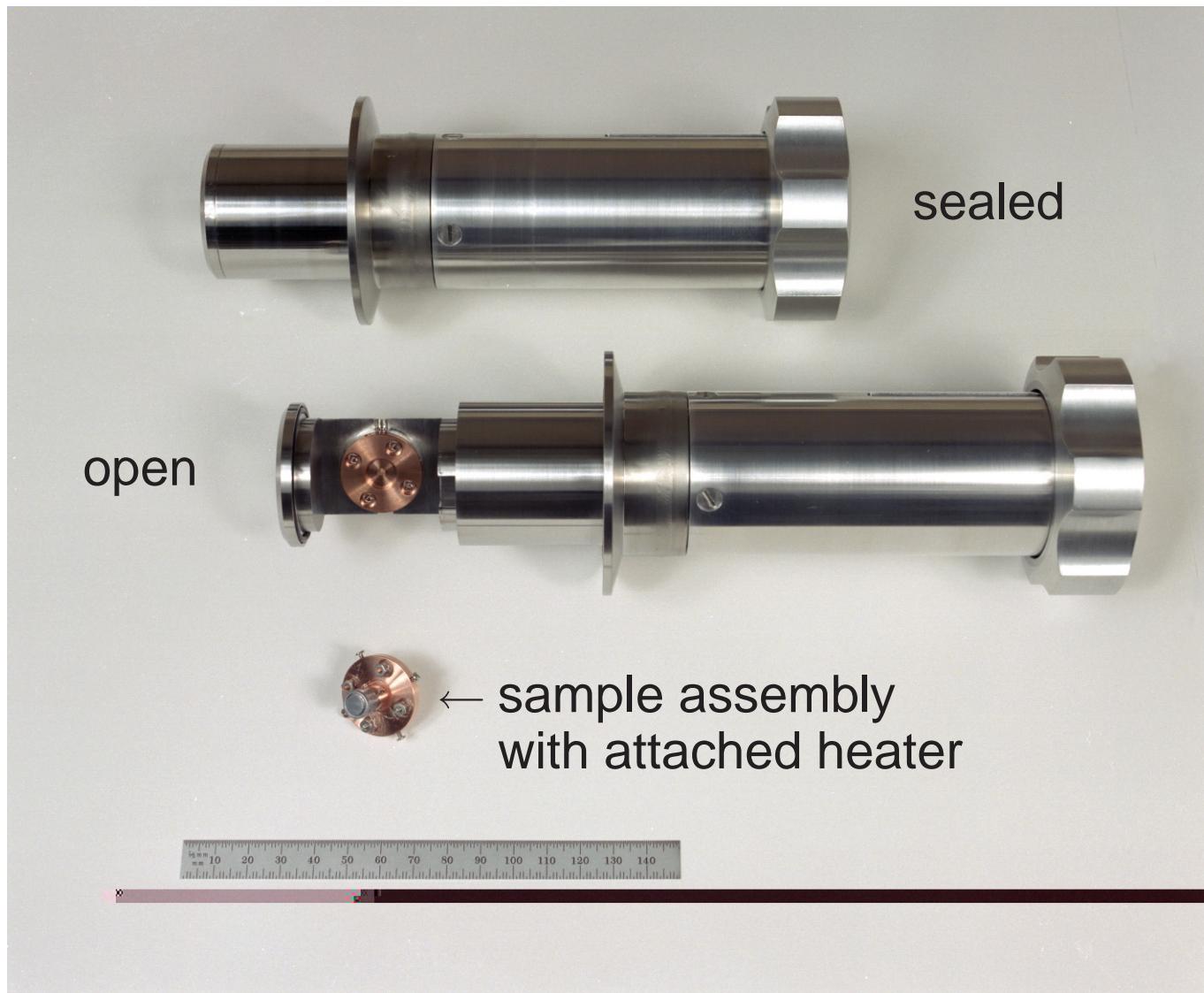


Surface analysis of liquid lithium

- Measurements have been made to determine the surface composition of liquid lithium in vacuum.
- High-purity lithium foil (99.9 %) was installed on a heated manipulator inside the ARIES UHV analysis chamber. All handling was done without air exposure.
- Low-energy ion scattering spectroscopy was used to measure the surface composition. The ion probe also served to sputter clean the surface.
- Measurements were made from 25 to 350 °C. Changes in the surface composition were monitored near the liquid/solid phase boundary.



A transfer device is used to move Li samples from glove boxes to the UHV analysis chamber.



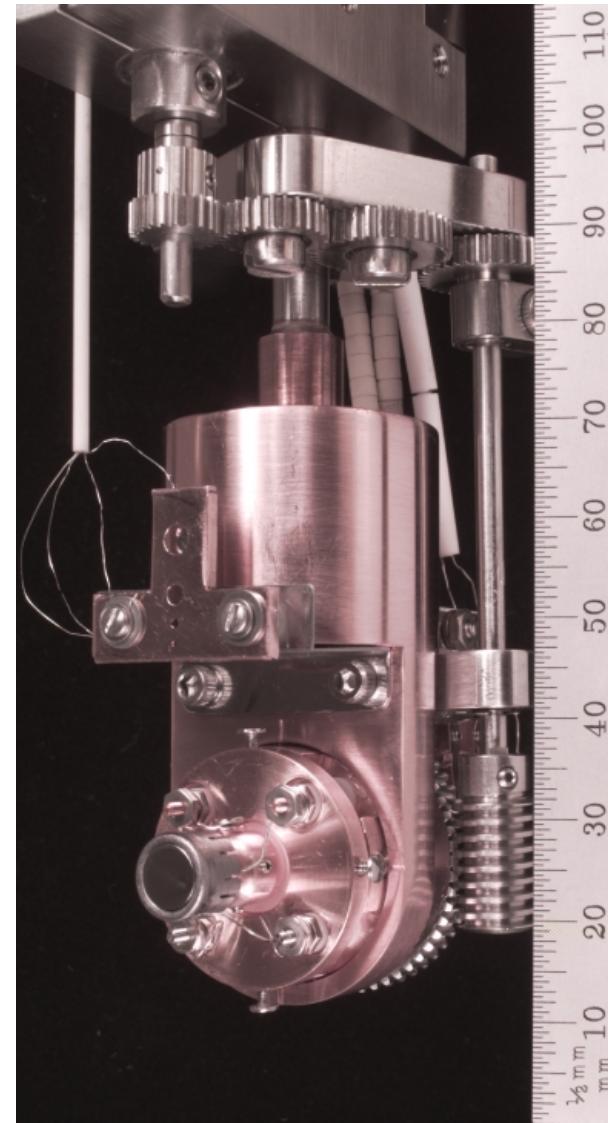
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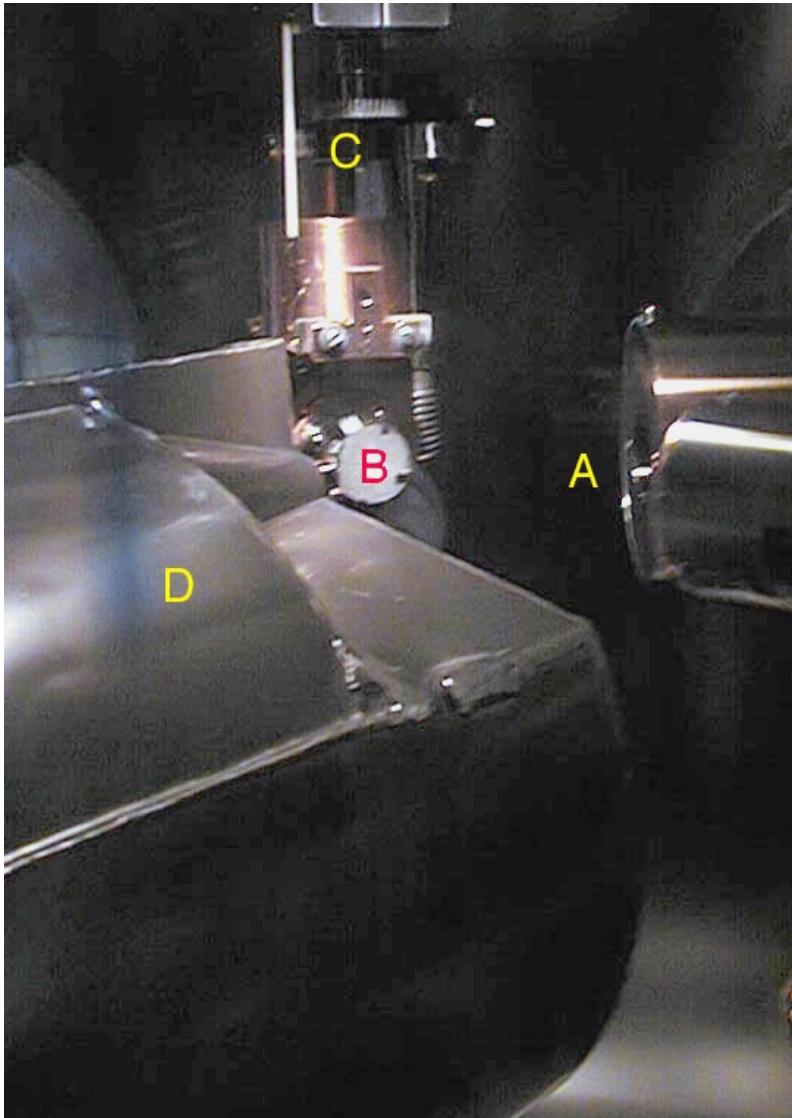


A UHV manipulator enables samples to be remotely inserted and examined.

- five axes of motion (x,y,z,α,ϕ)
- temperature control from -150 to 1000 °C
- electrical isolation
- built-in Faraday cups
- vacuum sample transfer capability



View inside UHV analysis chamber



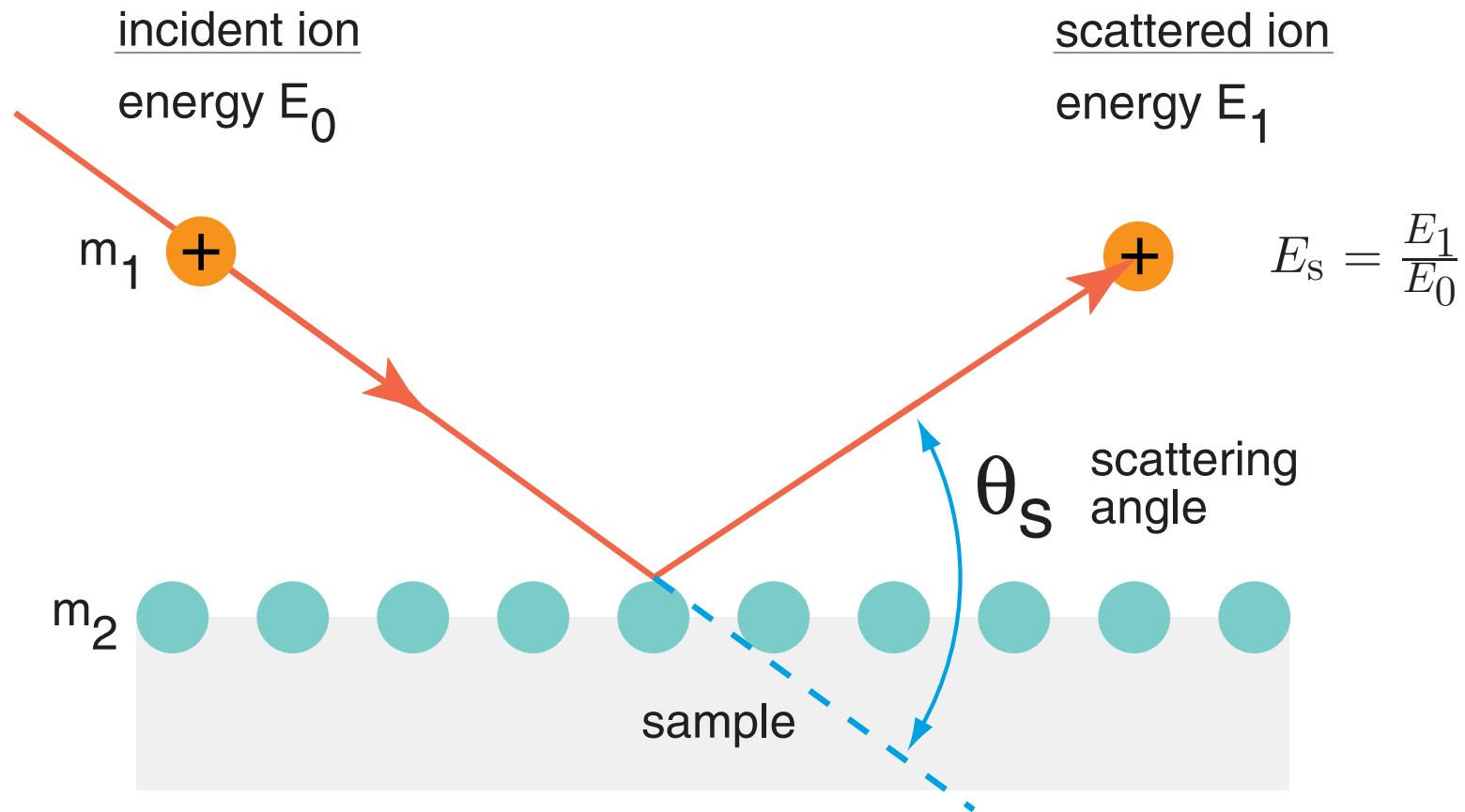
- A ion-beam gun
- B sample
- C sample manipulator
- D energy analyzer

sample diameter: 2.5 cm

UHV: ultra-high vacuum
(base pressure < 50 nPa)



Surface measurements consist of aiming the ion beam at a surface and measuring the energy loss of reflected ions.



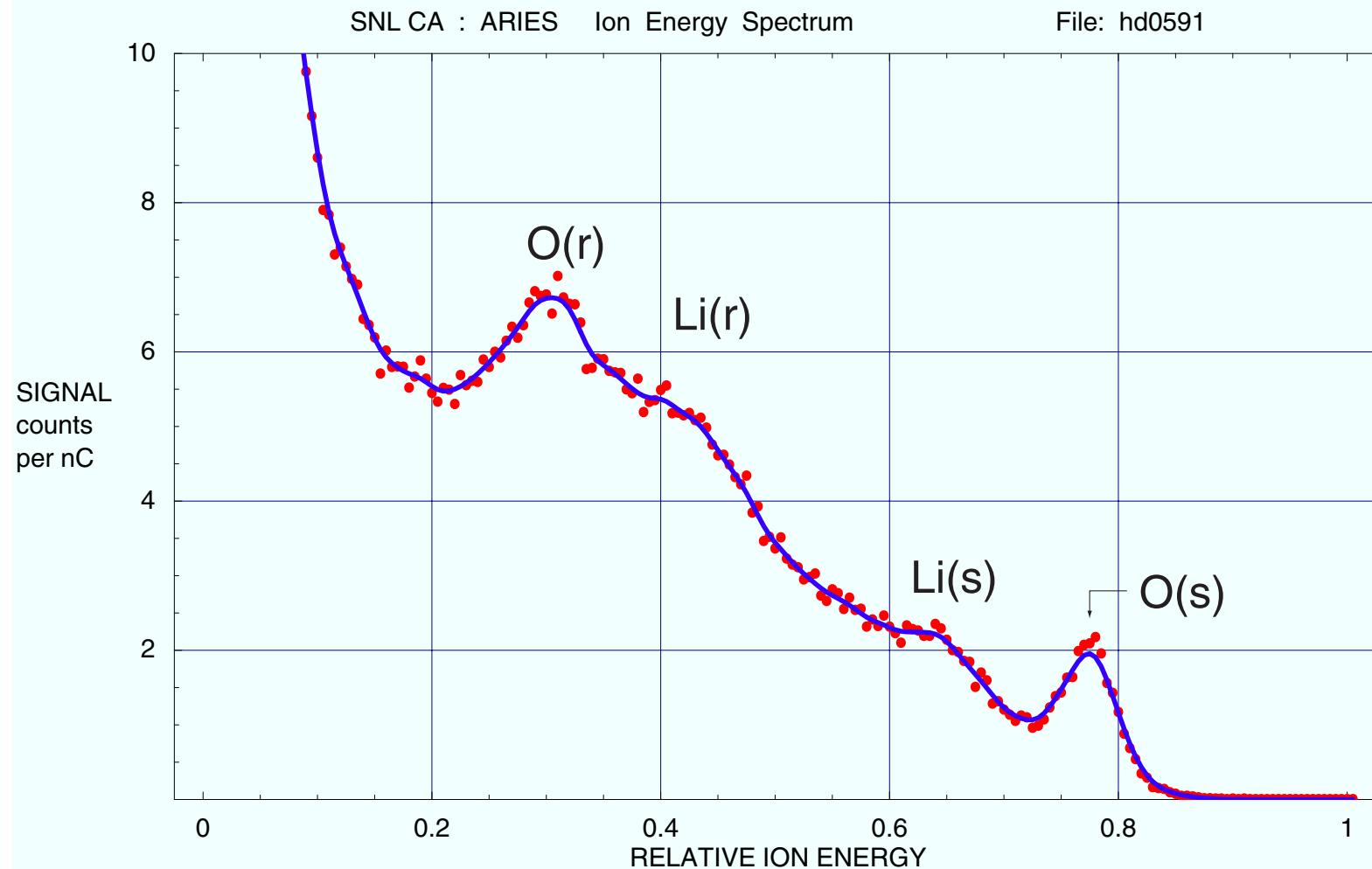
$$m_2 = m_1 \frac{1 + E_S - 2\sqrt{E_S} \cos \theta_S}{1 - E_S}$$



Initially oxygen covers much of the surface.

500 eV He+ --> Li foil

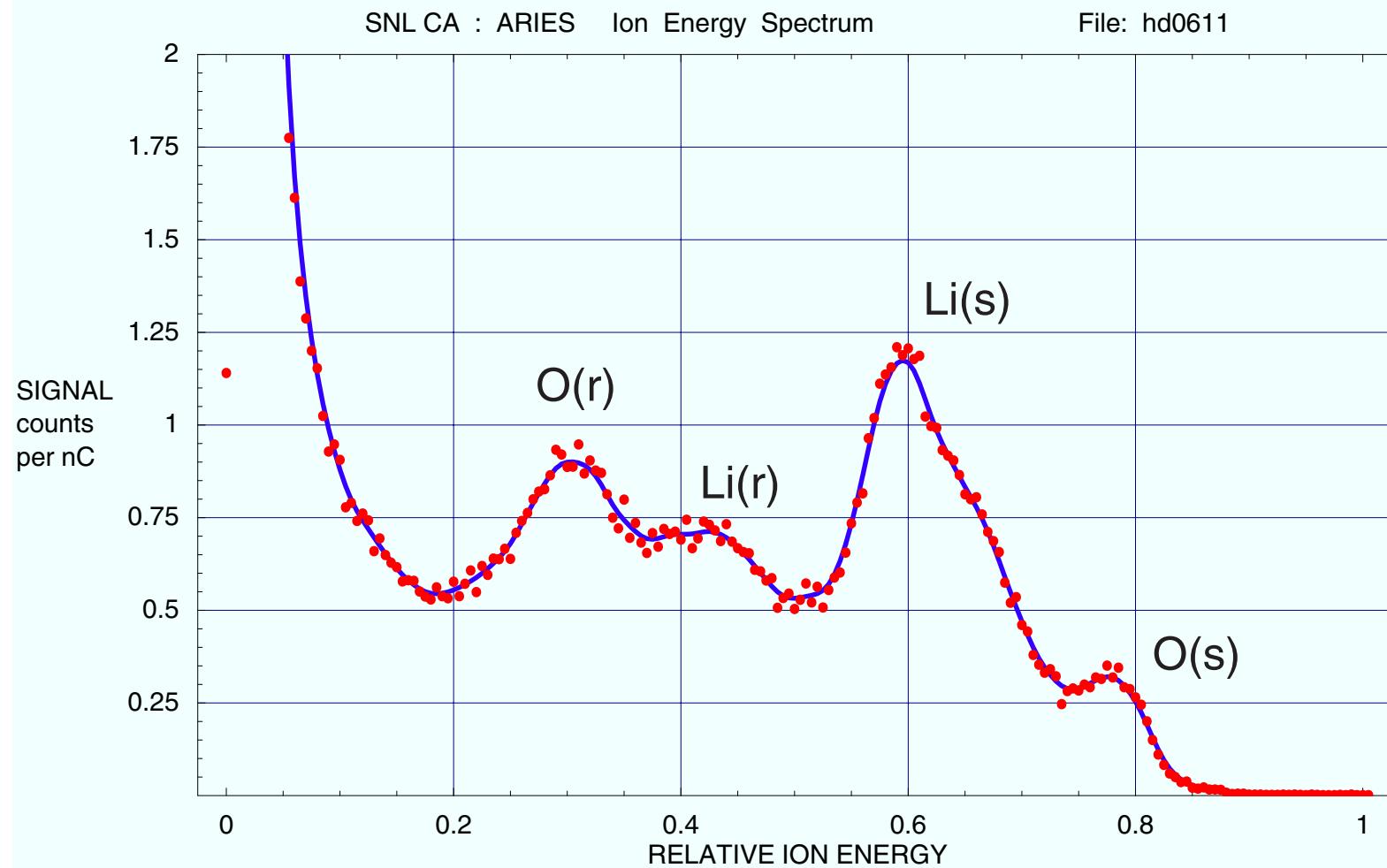
Theta = 45.0 deg Alpha = 75.0 deg Temp = 25 C



Sputter cleaning exposes the Li substrate.

500 eV He+ \rightarrow Li foil

Theta = 45.0 deg Alpha = 67.5 deg Temp = 29 C



Clean lithium sample: solid and liquid



solid

$\approx 170 \text{ }^{\circ}\text{C}$



liquid

$\approx 190 \text{ }^{\circ}\text{C}$



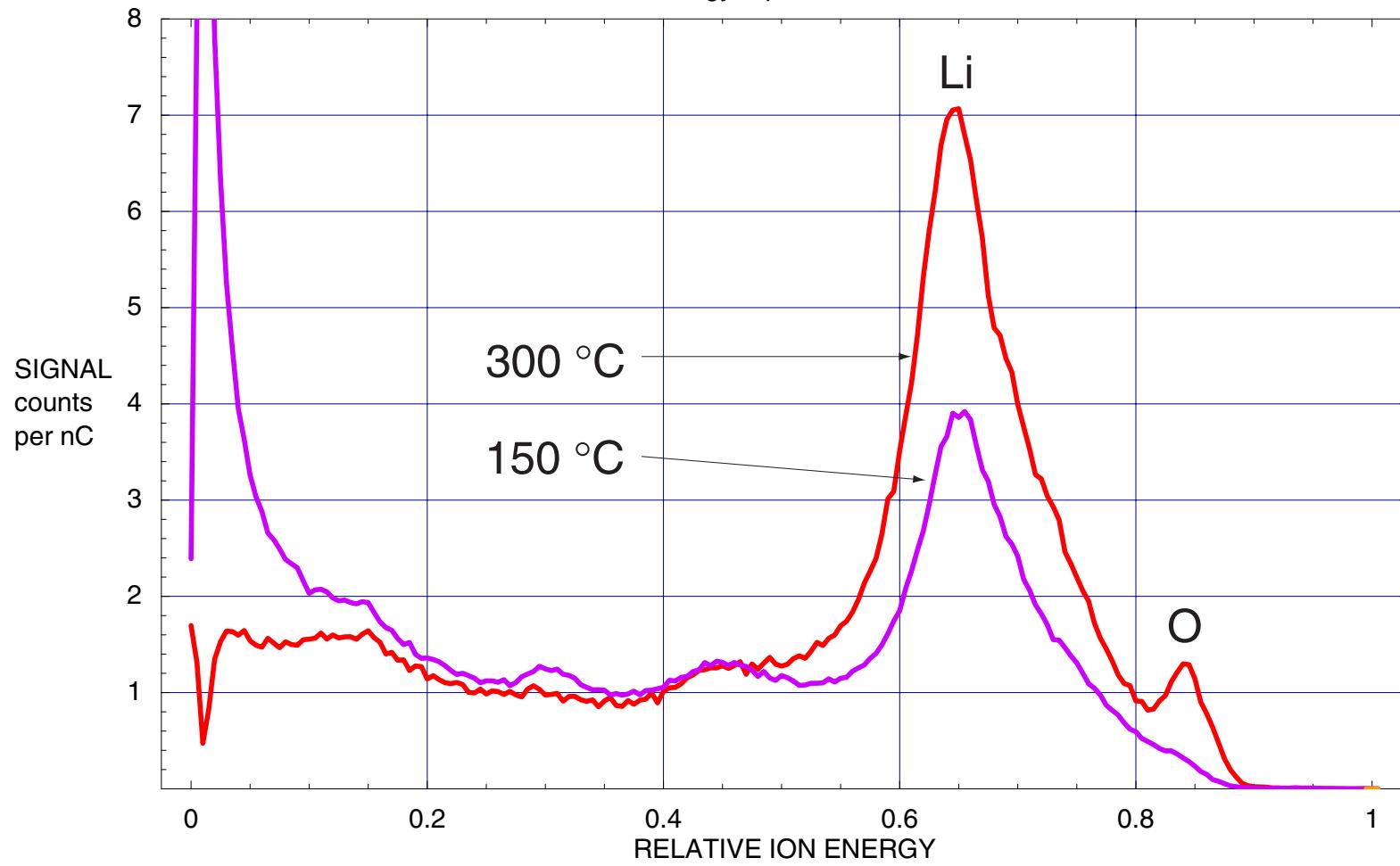
Surface oxygen increases upon melting.

1000 eV He+ \rightarrow Lithium

Theta = 45.0 deg Alpha = 67.5 deg Temp = 150, 300 C

SNL CA : ARIES Ion Energy Spectrum

Files: hd0727,hd07



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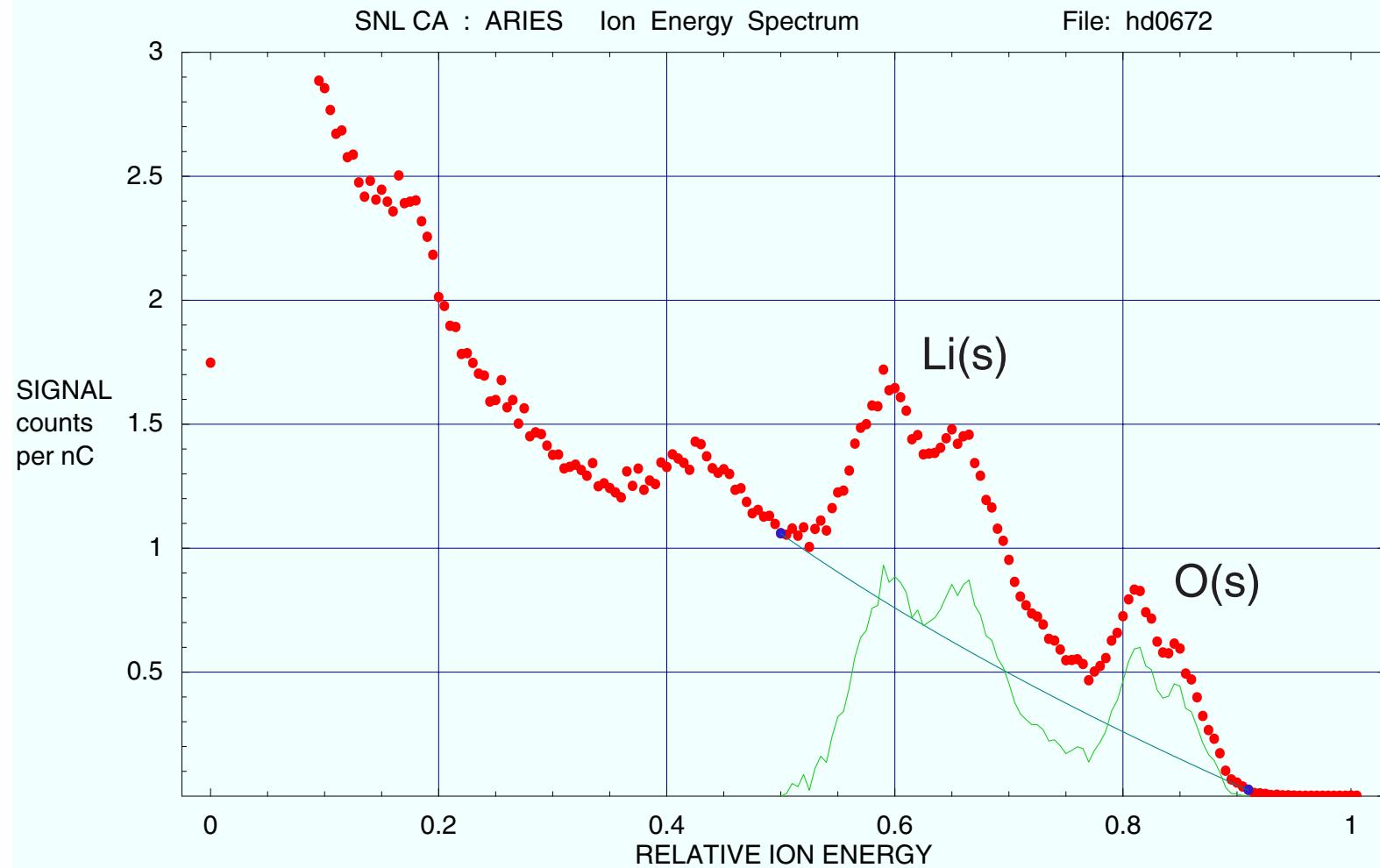
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At 350 °C, surface O is still present.

500 eV He+ \rightarrow Li liquid

Theta = 45.0 deg Alpha = 67.5 deg Temp = 350 C



Scattering calculations for 500 eV He → Li

Differential Collision Cross Sections for He -> Li

E0 (keV) = 0.500 Z1 = 2 M1 (amu) = 4.003 Z2 = 3 M2 (amu) = 6.941

Scattering formula: Gauss-Mehler quadrature

Potential function: ZBL

Screening function: ZBL

Screening length = 0.19042 Å

Reduced energy = 0.69893

Lab Angle (degrees)	Relative Angle (degrees)	Impact Parameter (Å)	Apsis (Å)	Lab Final Energy (eV)	Lab Cross Section (Å²/sr)	Event Type
15.00	23.58	0.2724	0.3124	4.8062E+02	7.2828E-01	S
	150.00	0.0220	0.1407	4.3289E+02	7.4182E-03	R
30.00	46.76	0.1614	0.2202	4.2694E+02	8.5759E-02	S
	120.00	0.0467	0.1480	3.4797E+02	9.7496E-03	R
45.00	69.07	0.1097	0.1827	3.5090E+02	2.2900E-02	S
	90.00	0.0785	0.1633	2.3198E+02	1.6039E-02	R
60.00	89.96	0.0786	0.1634	2.6816E+02	8.7368E-03	S
	60.00	0.1275	0.1950	1.1599E+02	3.6390E-02	R
75.00	108.85	0.0574	0.1525	1.9305E+02	4.0459E-03	S
	30.00	0.2302	0.2761	3.1080E+01	1.5309E-01	R
90.00	125.22	0.0421	0.1463	1.3423E+02	2.1075E-03	S



Scattering calculations for 500 eV He → O

Differential Collision Cross Sections for He -> O

E0 (keV) = 0.500 Z1 = 2 M1 (amu) = 4.003 Z2 = 8 M2 (amu) = 15.999

Scattering formula: Gauss-Mehler quadrature

Potential function: ZBL

Screening function: ZBL

Screening length = 0.16816 Å

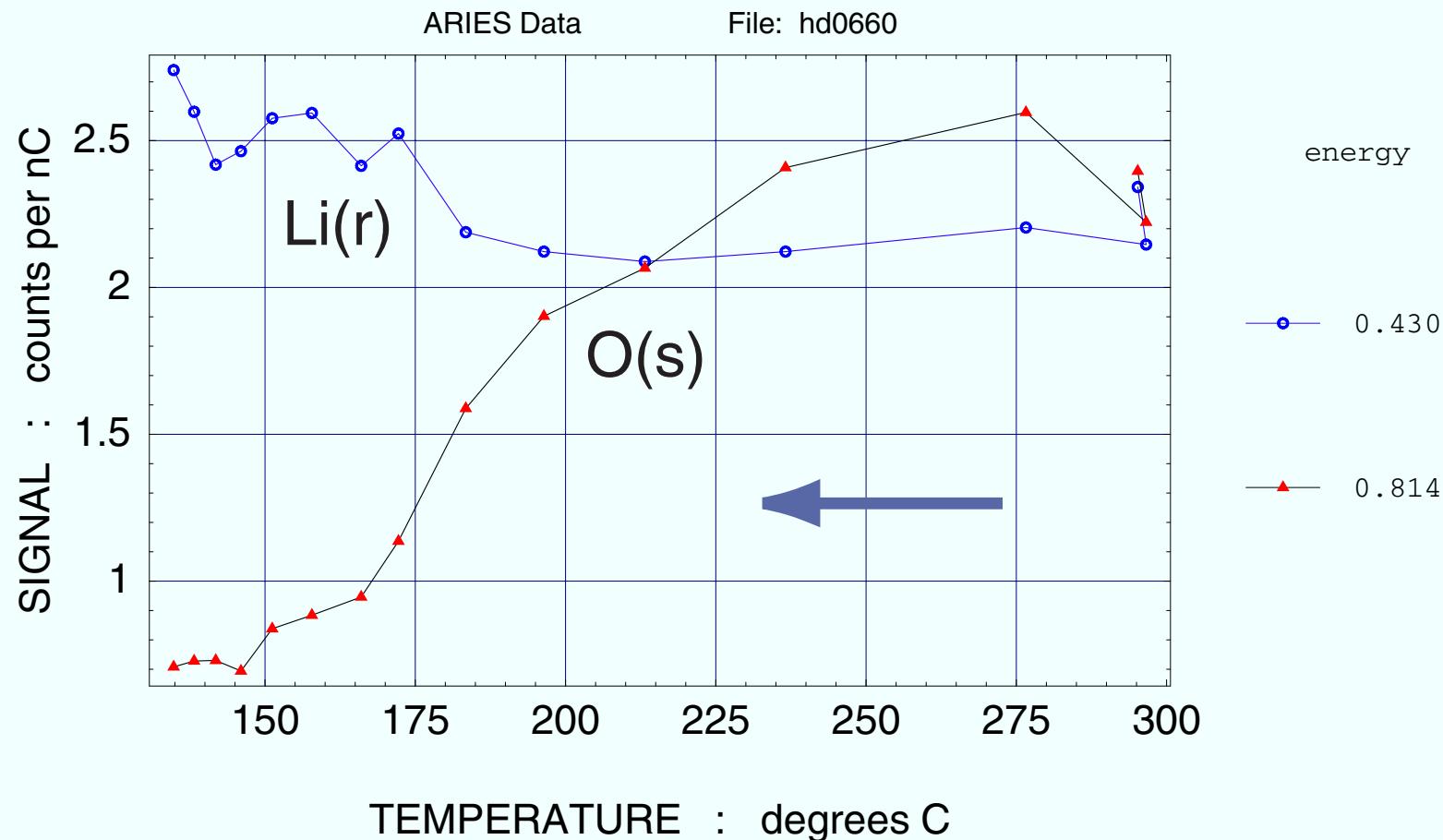
Reduced energy = 0.29189

Lab Angle (degrees)	Relative Angle (degrees)	Impact Parameter (Å)	Apsis (Å)	Lab Final Energy (eV)	Lab Cross Section (A*Å/sr)	Event Type
15.00	18.71	0.4278	0.4710	4.9154E+02	1.4471E+00	S
	150.00	0.0343	0.2091	2.9871E+02	1.8011E-02	R
30.00	37.19	0.2804	0.3483	4.6745E+02	2.0967E-01	S
	120.00	0.0725	0.2194	2.4012E+02	2.3074E-02	R
45.00	55.19	0.2051	0.2927	4.3130E+02	6.4719E-02	S
	90.00	0.1202	0.2404	1.6008E+02	3.6131E-02	R
60.00	72.51	0.1568	0.2611	3.8802E+02	2.7826E-02	S
	60.00	0.1901	0.2824	8.0039E+01	7.5021E-02	R
75.00	88.99	0.1221	0.2414	3.4276E+02	1.4511E-02	S
	30.00	0.3242	0.3833	2.1446E+01	2.6760E-01	R
90.00	104.49	0.0954	0.2285	2.9987E+02	8.6571E-03	S

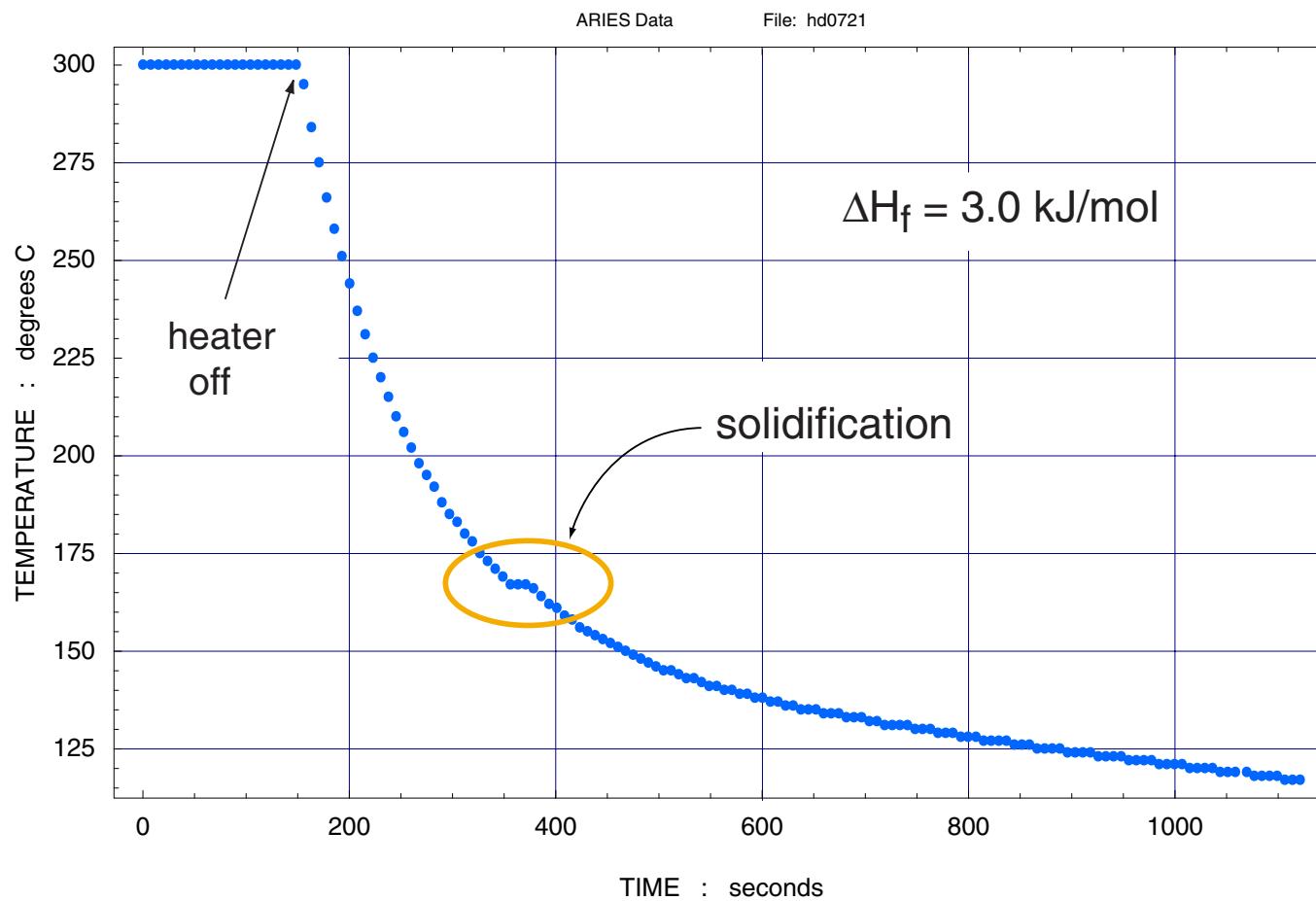


Oxygen level drops near the phase boundary.

500 eV He \rightarrow Lithium
cooling across liquid/solid phase boundary
 $\theta = 45.0$ deg $\alpha = 67.5$ deg



The solidification point is evident from the temperature profile during cooling.



Surface analysis of liquid lithium – summary

1. The sample was sputter cleaned in UHV ($<10^{-9}$ torr) using 500 eV He ions to remove the native oxide.
2. Upon melting, the surface becomes smooth and its reflectivity increases. The liquid evaporates quickly above 400 °C.
3. The predominant species observed at the liquid surface are lithium and oxygen. An estimate of the surface composition at 350 °C gives about 10% O coverage of the Li surface.
4. No evidence of high mass impurities has been found on the liquid Li surface.
5. Surface oxygen coverage increases upon melting. The surface/bulk segregation ratio is about 10^2 .



Surface analysis of a liquid tin-lithium alloy

- Sn-Li alloys have potential advantages:
 - > lower vapor pressure
 - > lower T inventory
 - > reduced reactivity.
- $\text{Sn}_{0.8}\text{Li}_{0.2}$ was prepared in-situ by melting Sn and Li foils.
- Low-energy ion scattering spectroscopy was used to measure the surface composition. The ion probe also served to sputter clean the surface.
- Measurements were made at temperatures up to 800 °C, where the material is a single-phase binary liquid.

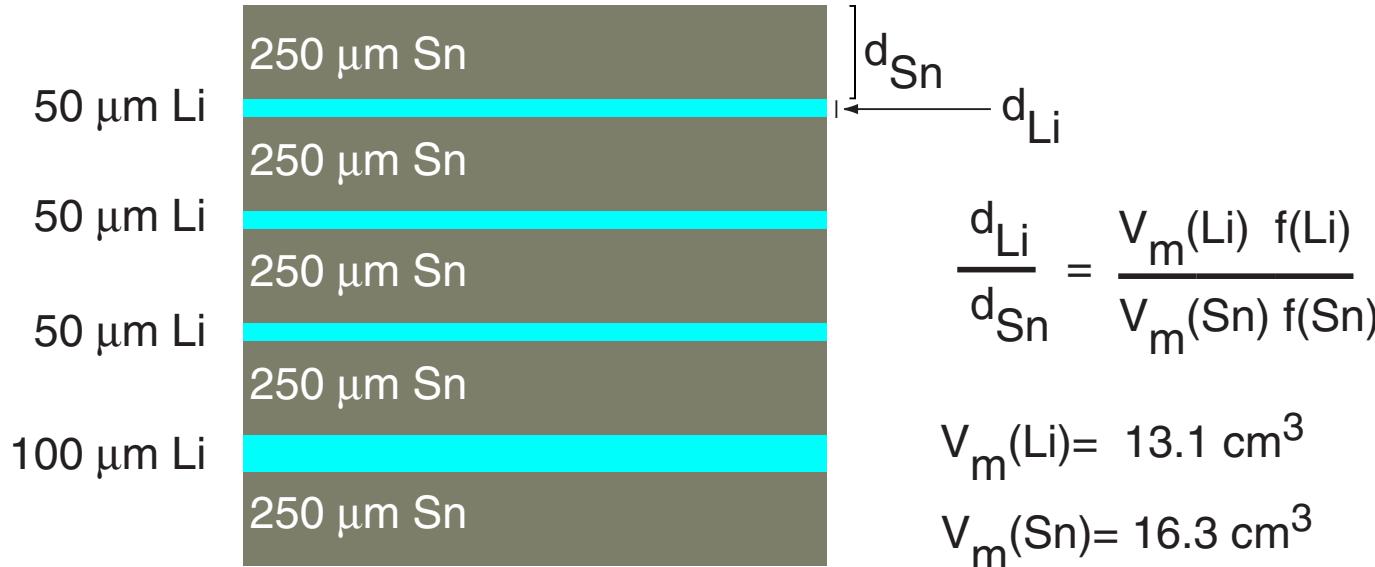


Melting points and densities of some Li and Sn containing materials

<u>material</u>	<u>melting point (°C)</u>	<u>density (g/cm³)</u>
SnH_4	-146	
Li	180.5	0.534
Sn_4Li	222	
Sn	231.93	7.265
LiH	688.7	0.78
Li_7Sn_2	783	
Li_2O	1570	2.013
SnO_2	1630	6.85



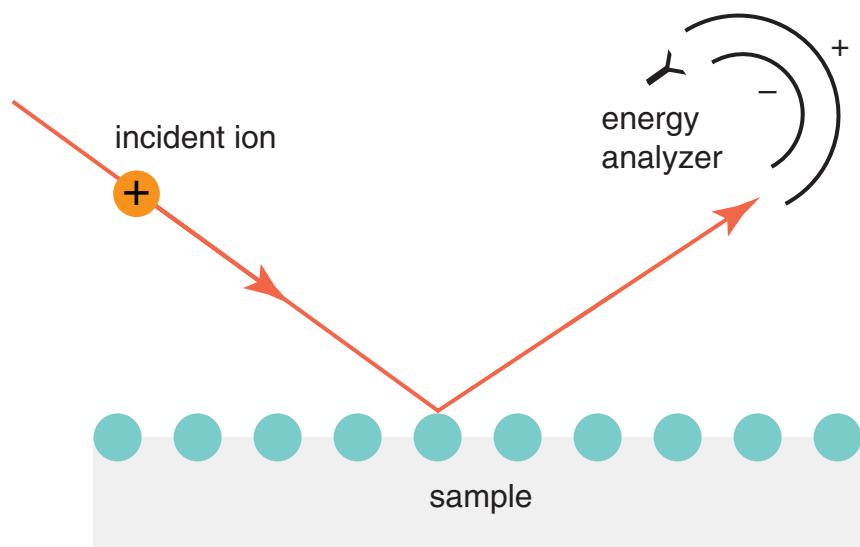
In-situ preparation of liquid 80% Sn - 20% Li alloy



- Layers of high purity Sn and Li foils are melted in vacuum.
- A 5:1 thickness ratio produces the desired stoichiometry.
- The top and bottom layers are Sn to reduce Li evaporation.



Parameters for ion scattering measurements of Sn-Li surface composition



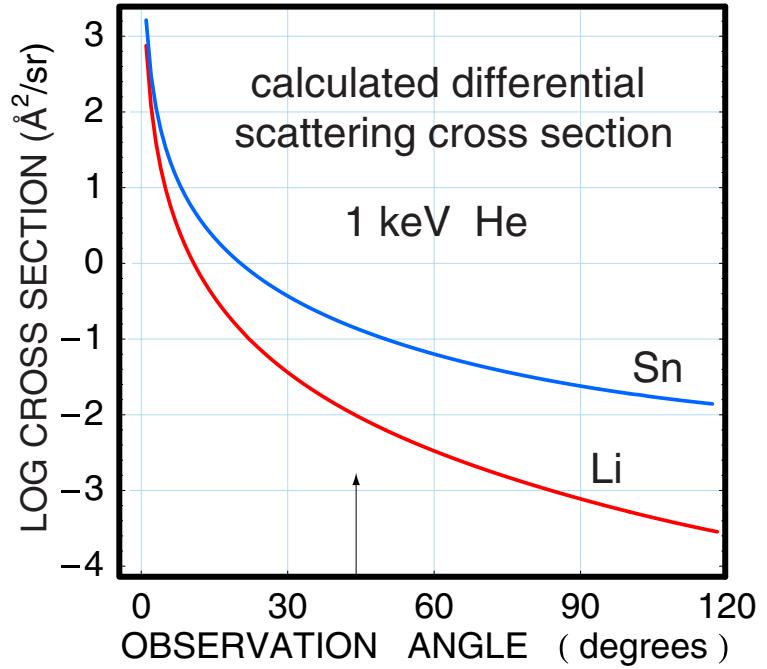
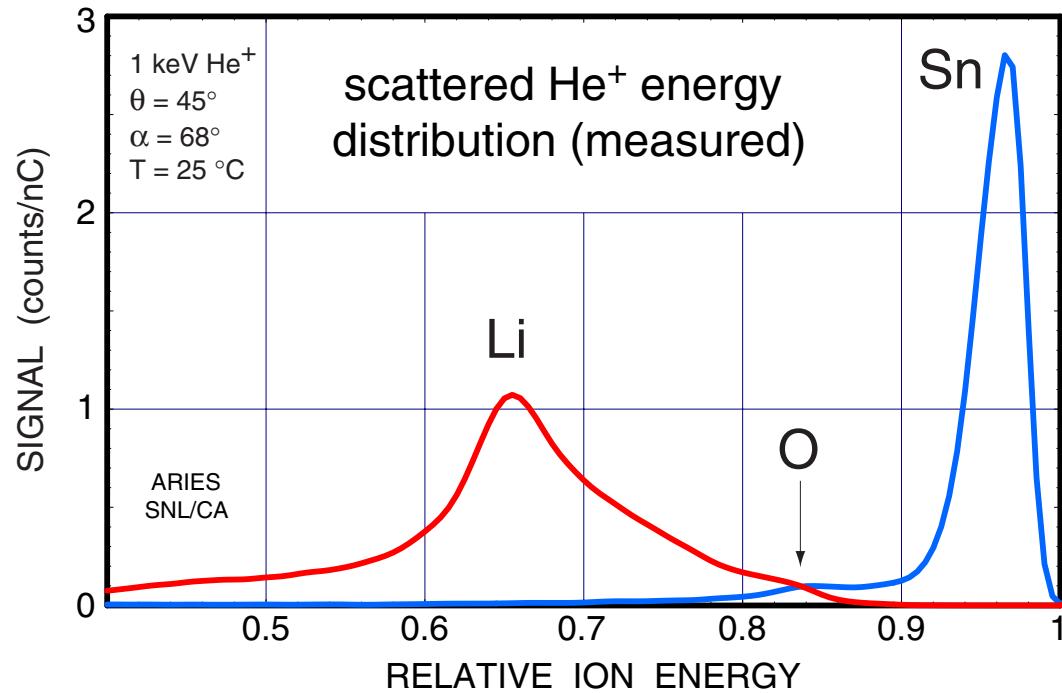
$$S \propto \rho \sigma$$

* for 1 keV He at
 45° scattering angle

<u>atom</u>	ρ <u>element density</u> <u>atoms/\AA^2</u>	σ <u>scattering CX</u> <u>($\text{\AA}^2/\text{sr}$)*</u>
Li	0.128	0.00907
O	0.123	0.0309
Sn	0.111	0.131



Measurements of forward scattered He⁺ give strong, well-resolved Sn and Li signals.



- 45° is good balance between mass resolution and signal strength.
- Surface oxygen can also be detected at this observing angle.
- Scattering from Sn more intense than from Li.

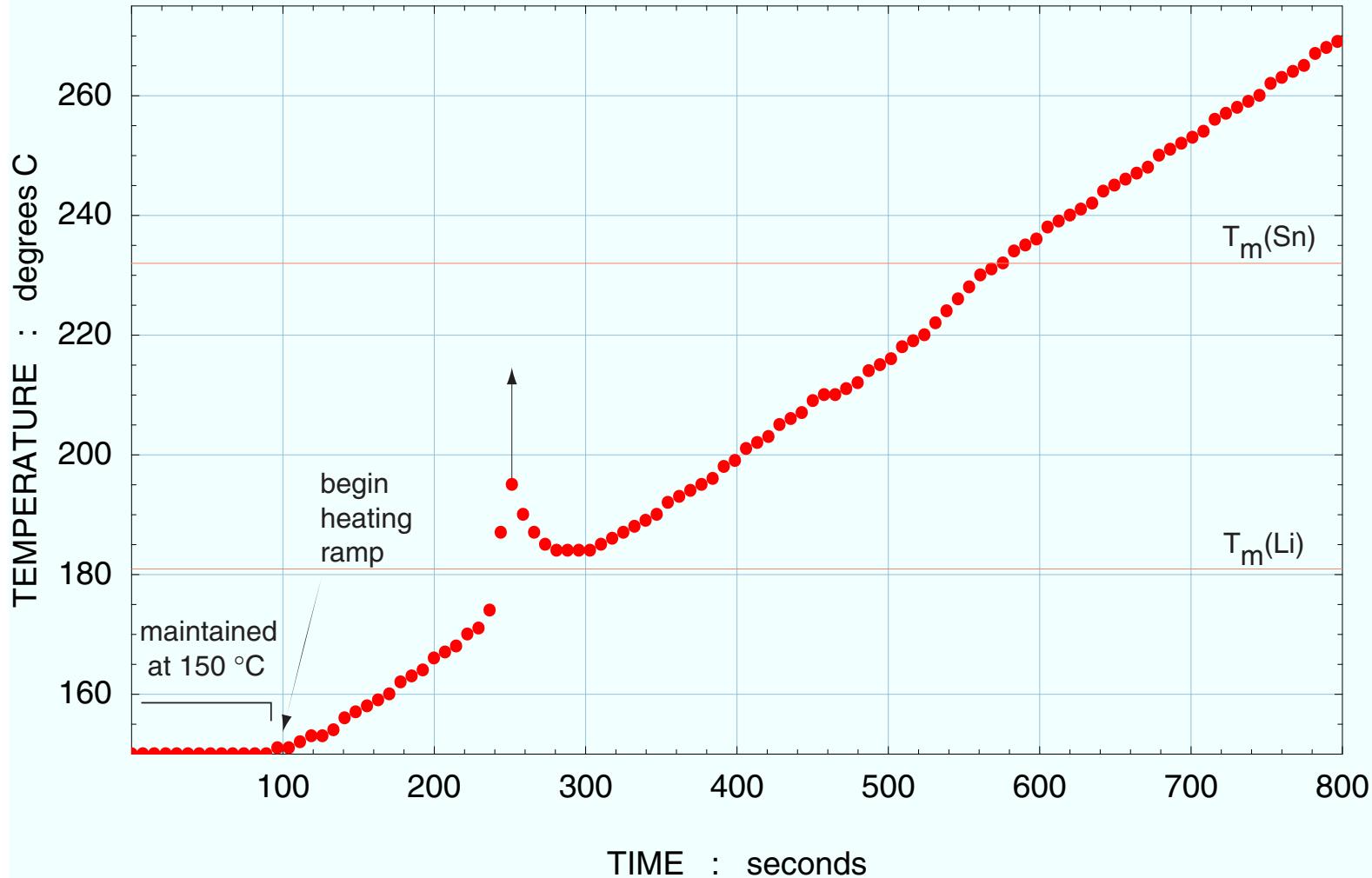


An exothermic reaction occurs near $T_m(\text{Li})$

first heating: Sn(0.25 mm) – Li(0.10 mm) – Sn(0.25 mm)

ARIES Data

File: hd0778



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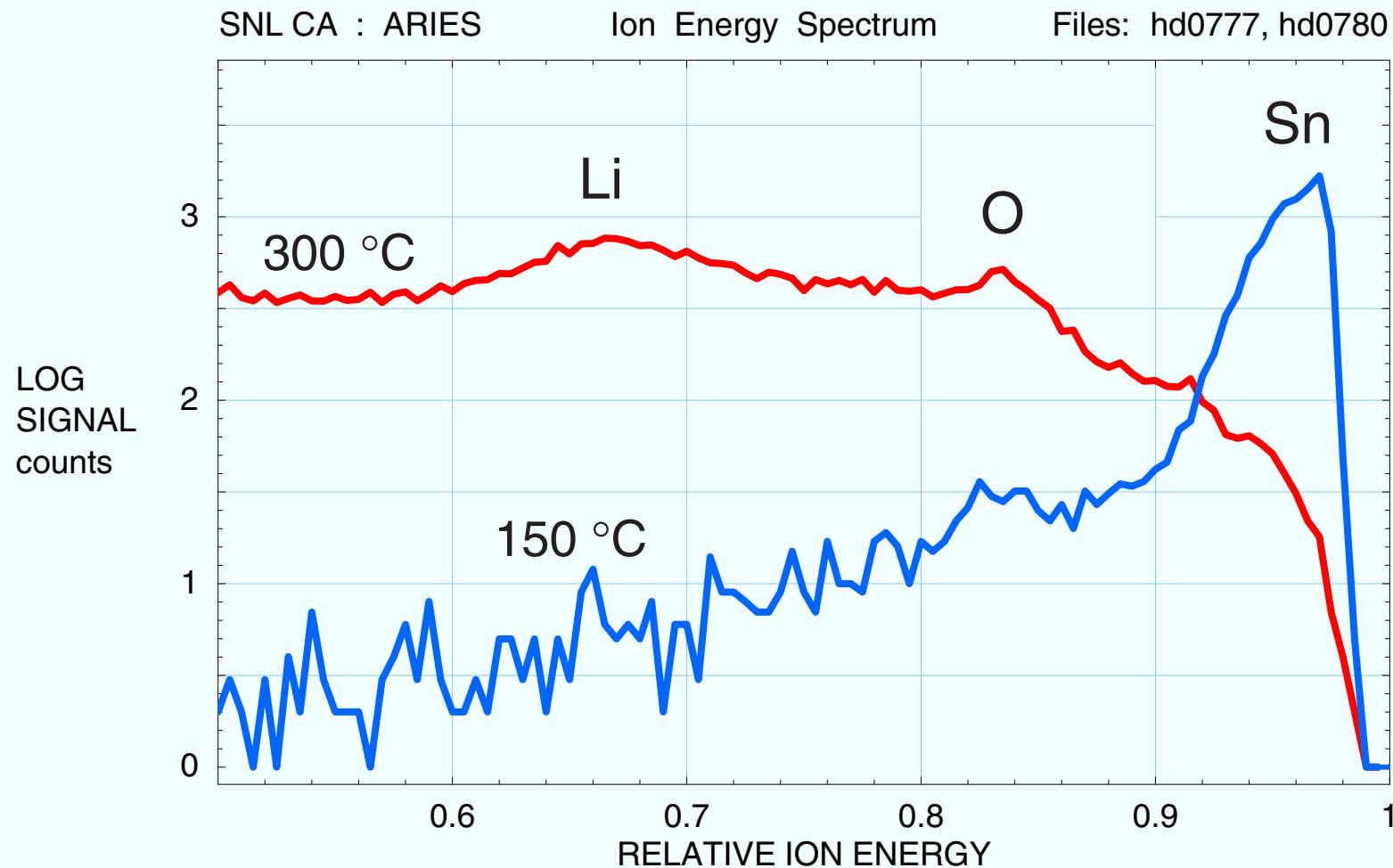


Upon melting, Li segregates to the alloy surface

1000 eV He⁺ ---> Sn(0.25 mm) – Li(0.10 mm) – Sn(0.25 mm)

first heating: below and above melting point

Theta = 45.0 deg Alpha = 67.5 deg Temp = 150, 300 C



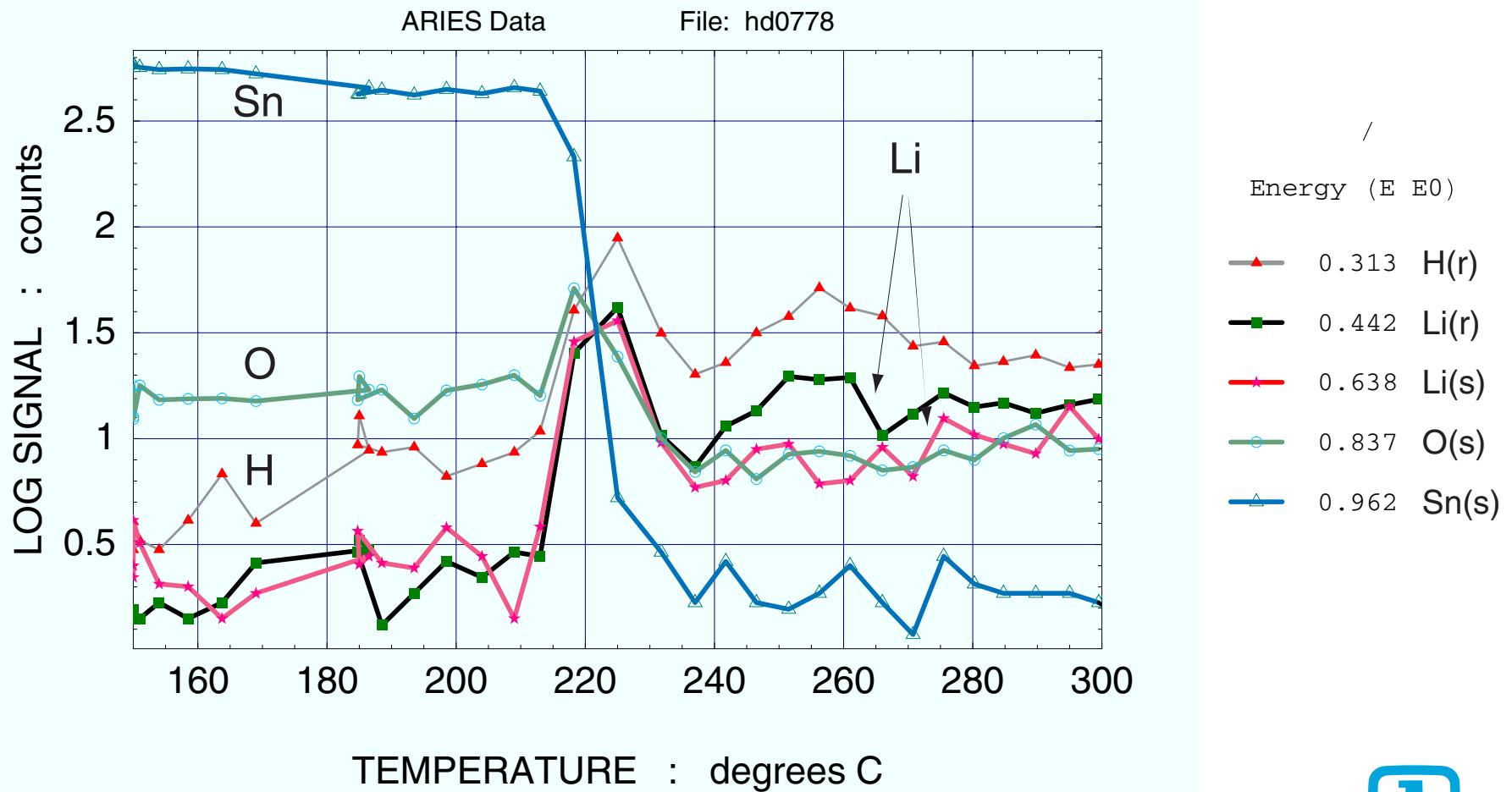
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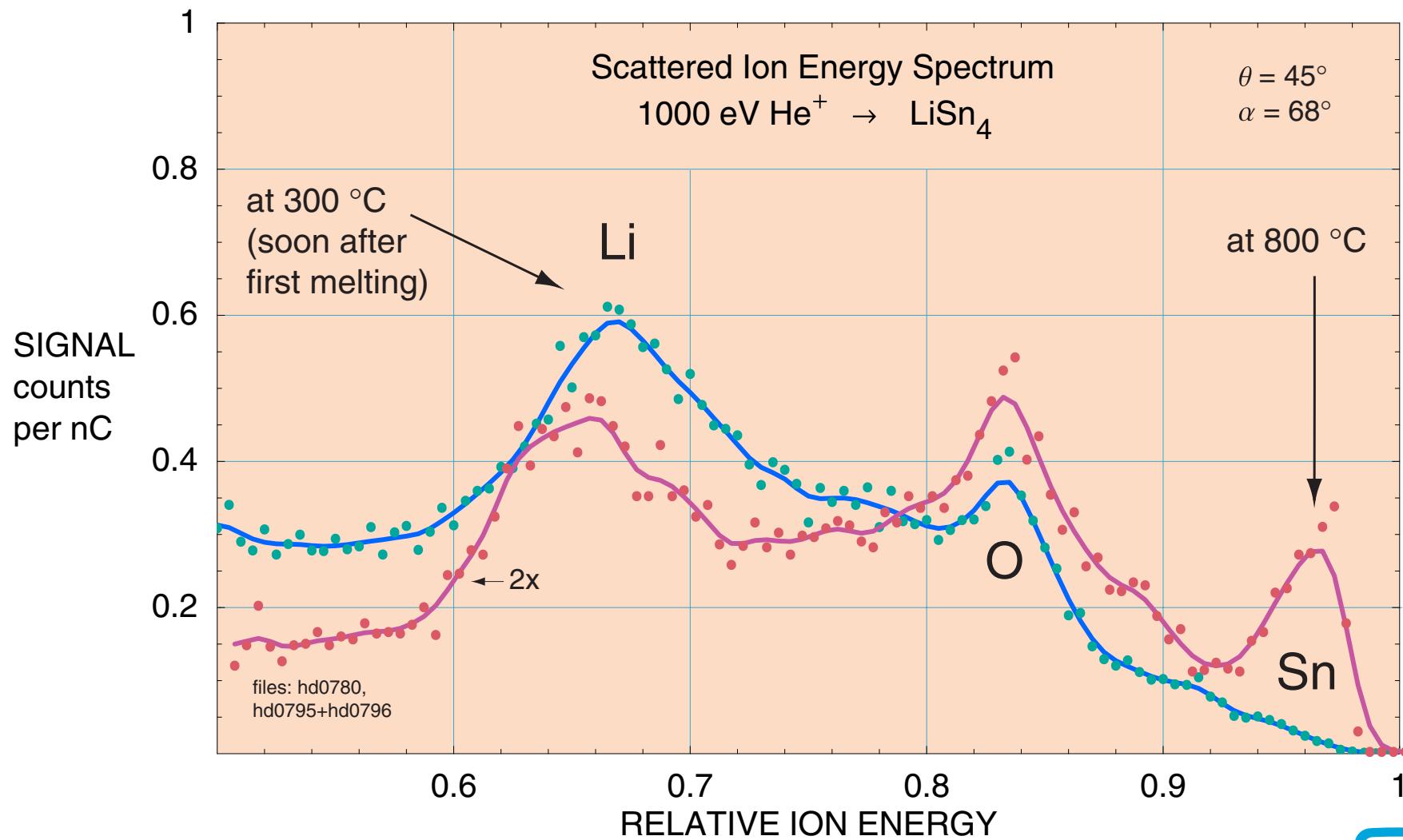
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Surface composition changes near $T_m(\text{Sn}_4\text{Li})$

first heating: Sn(0.25 mm) – Li(0.10 mm) – Sn(0.25 mm)



At higher temperatures, Sn appears.



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Surface analysis of liquid Sn_4Li – summary

1. An exothermic reaction between elemental Li and Sn occurs near $T_m(\text{Li})$ and produces a liquid Sn-Li alloy.
 2. Upon melting, the alloy becomes highly enriched in Li at the surface.
 3. Oxygen also segregates to the liquid Sn-Li alloy surface.
 4. At high temperatures (800°C), Li depletion and Sn enrichment observed at the surface.
- ⇒ Li is the plasma-facing material in liquid Sn-Li, at least initially.



Planned work

- Segregation rate measurements.
 - measurements will determine the kinetics of:
 - O transport to liquid Li surfaces,
 - Li transport to liquid Sn-Li surfaces,
 - O transport to liquid Sn-Li surfaces.
 - the TRIDYN code will be used to evaluate the sputter flux composition.
- Li sputtering measurements on Li and Sn-Li.
 - the variation in yield with angle-of-incidence will be measured to determine under what conditions the yield may exceed unity.

